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# The Sources of Radon to the Radioactive Springs

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## §1. Introduction.

Most of the radioactive springs contain such small quantities of radium compared with their considerable amounts of radon that the major part of radon in them must originate from other sources in the ground, and with respect to these sources, following two conditions are considered.

- (1) Although a large amount of radon has its origin in that of radium dissolving in the deep groundwater, only radon is left in the water after the greater part of radium was deposited until the outlet.
- (2) Owing to the larger dissolution velocity of radon than radium, the flowing groundwater dissolves only radon produced in the surrounding rocks, so that its mother substance, that is radium, is left as it was.

The second condition is now rather recognized by many researchers than the first which is considered to be doubtful from the observations in many spring areas showing that the higher content of radium generally follows the lower content of radon. Then, it has been expressed that the sinters of radioactive springs played the most important parts to supply the spring waters with their large amounts of radon.

Kuroda introduced the hypothetical information of underground radon-supply from the opposed relation between the contents of radium and radon in Masutomi Springs. Iwasaki<sup>2)</sup> has made some experiments to take knowledge of the radon dissolution velocities from spring sinters. Laboratory investigations for the supply of radon to the flowing water were made by Mache<sup>3)</sup> and Spitzuin<sup>4)</sup>. Analytical study on the underground vertical distribution of radon was attempted by G. Aeckerlein<sup>5)</sup> using the data obtained during the period of boring. However, the quantitative research has not been undertaken for the sources of radon to the radioactive springs.

In this paper, theoretical equations are presented which describe the relation between the contents of radon in the spring water and radium in the mother rocks and which permit the calculation of the total amount of radium required to support any radioactive spring.

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## §2, Radon-supply from rocks to the groundwater.

Notations are defined as follows.

- $\rho$  ; Rn content of groundwater. curie/cc.  
 $\rho_r$  ; Ra content of rocks. g/cc.  
 $h$  ; The ratio between radon in curie and radium in gram under radioactive equilibrium. 1 curie/g.  
 $q$  ; Flow of groundwater. cc/min.  
 $v$  ; Velocity of groundwater. cm/min.  
 $\lambda$  ; Coefficient of disintegration of radon.  $1.258 \times 10^{-4} \text{min}^{-1}$ .  
 $x$  ; The length along the running course of groundwater toward the outlet. cm.  
 $D$  ; Diffusion coefficient of radon through the water.  
 $\alpha$  ; Solubility of radon.  
 $A$  ; Contact area between water and rocks per unit length along the flow of groundwater.  $\text{cm}^2$ .  
 $S$  ; The sectional area of the groundwater.  $\text{cm}^2$ .

(a) Radon gas produced from underground radium dissolves into the groundwater after diffusing through the rocks. The dissolution velocity of radon per unit length along the course of groundwater may be in proportion to the contact area and to the difference between radon concentrations in the undisturbed rocks and in the water, as represented by  $CA \left( h \rho_r - \frac{1}{\alpha} \rho \right)$  where  $C$  is assumed to be constant.

Simplifying concepts are taken as the uniform underground condition of constant  $q$ ,  $v$ ,  $A$ ,  $S$  and  $\rho_r$ .

We can find the next equation to show the change in the amount of radon transported with the flow of groundwater.

$$\frac{\partial q \rho}{\partial t} = CA v \left( h \rho_r - \frac{1}{\alpha} \rho \right) - \lambda \rho q + Dq \frac{\partial^2 \rho}{\partial x^2} - v \frac{\partial q \rho}{\partial x}$$

Supposing the steady state in which  $\frac{\partial \rho}{\partial t} = 0$  and that the diffusing of radon may be negligible in the water.

$$\frac{\partial \rho}{\partial x} = - \left( \frac{\lambda}{v} + \frac{CA}{\alpha q} \right) \left( \rho - \frac{\alpha v CA h}{\alpha \lambda q + v CA} \rho_r \right)$$

Taking that  $\rho = 0$  at  $x = 0$  and since  $q = vS$ ,

$$\rho = \frac{\alpha CA h}{\alpha S \lambda + CA} \rho_r \left( 1 - e^{-\frac{\alpha q \lambda + CA v}{\alpha v q} x} \right)$$

Radon content in the water after flowing for  $T$  min. from the position at  $x = 0$  is given as follows.

$$\rho = \frac{\alpha CAh}{\alpha S\lambda + CA} \rho_\gamma \left\{ 1 - e^{-\left(\lambda + \frac{CA}{\alpha S}\right)T} \right\} \quad (1)$$

One of the methods for finding the value of  $C$  involved in equ. (1) is to make the laboratory experiment concerning the dissolution of radon to water from rocks. When the rock of known radon content is put in a closed vessel filled up by the water, the variation in time of radon content assumed to be uniform in the water can be given by the next equation.

$$V \frac{\partial \rho}{\partial t} = CA \left( h\rho_\gamma - \frac{1}{\alpha} \rho \right) - \lambda \rho V$$

where  $V$  is the volume of water and  $A$  is the contact area between water and rock.

Since  $\frac{\partial \rho}{\partial t}$  becomes nearly equal to zero after sufficiently long time.

$$\rho = \frac{\alpha CAh}{CA + \alpha \lambda V} \rho_\gamma$$

$$\frac{h\rho_\gamma}{\rho} = \frac{1}{\alpha} + \frac{\lambda V}{CA} = K$$

$$\therefore C = \frac{\alpha \lambda V}{A(\alpha K - 1)}$$

$1/K$  may be called as the coefficient of radon dissolution from the rock, the value of which is found, for example, to range from 0.03 to 0.09 for spring sinters in Ikeda and equal about 0.02 for those in Yukakae by the calculations from the experimental data of Iwasaki. But the exact value of  $A$  is hardly known in practice and it may be impossible to say that the value of  $C$  thus obtained agrees with that under the actual state in the ground.

Accordingly, it is desirable to devise some other measurements in field or laboratory fitted to the actual condition.

(b) For the last treatment, rocks were considered to have uniform radium content, but it has been reported by Piggot and Kurbatov that the largely increasing concentrations of radium were found in the portion of rock being in contact with the ground-water. If there is greatly concentrated radium only in the thin layer on the contact surface, above-mentioned treatment can be more simplified as follows by assuming that all of radon produced from radium on the contact surface directly dissolve into the water.

When we assume that the  $R$  g of radium per unit length distributes uniformly along the running course of groundwater, the amount of radon produced per unit time with the decay of radium in the unit length is equal to  $\lambda hR$  (curie/min. ).

Then, under the steady condition,

$$q \frac{\partial \rho}{\partial x} = \lambda hR - \lambda \rho \frac{q}{v}$$

The solution satisfying the condition, that is  $\rho = 0$  at  $x = 0$ , is

$$\rho = \frac{vhR}{q} \left(1 - e^{-\frac{\lambda}{v}x}\right)$$

Radon content after flowing for the time interval of  $T$  min in such a region is

$$\rho = \frac{hR}{S} \left(1 - e^{-\lambda T}\right) \quad (2)$$

If above condition is satisfied over the distance of  $L$  cm along the course of groundwater through which the flow takes  $T$  min, the total amount of radium is represented by  $LR$  (g). Since  $qT = SL$ ,

$$\rho = \frac{LhR}{q} \cdot \frac{1 - e^{-\lambda T}}{T} \quad (3)$$

When the equ. (3) is applied to the case of (a), it is clear that  $LR$  is expressed as the total amounts of radium contributing to the radon in the groundwater.

### §3. Radon content at the outlet.

Radon content in any spring water is generally shown by the measured value at the outlet. This value, however, is often found to fluctuate in large range. The author previously explained the possibility of those fluctuations in Rn content as a result of the variation in the loss of radon<sup>8)</sup> diffusing through the surrounding soil air from flowing groundwater, especially, in the shallow region under the ground surface. Such amount of loss through unit area of water surface per unit time was presented as  $K \frac{\rho}{\alpha}$ , in which the value of  $K$  was varied with ground condition or the depth from ground surface. For example, it was given for 1.5 m depth, as  $3.32 \times 10^{-2} \text{ min}^{-1}$  under the free condition for radon-exhalation or as  $1.65 \times 10^{-2} \text{ min}^{-1}$  under the perfectly closed condition. Therefore, the effect of  $K$  near the surface must be entered into the present calculation when the value of  $\rho$

at  $t = T$  in the last section. is taken as at the outlet.

We assume that such loss of radon occurs only during the flow for the time interval of  $T'$  min until the outlet and  $K$  is taken as the value for the mean depth in such a region. Let  $a$  or  $b$  be the area in  $\text{cm}^2$  per unit length through which radon is given to or lost from the groundwater. The next relation may be always held.  $a + b = A$ .

Radon contents at the outlet are found by the following treatments in both cases of (a) and (b).

$$(a) \quad q \frac{\partial \rho}{\partial x} = Ca \left( h\rho_\gamma - \frac{1}{\alpha} \rho \right) - Kb \frac{a}{\alpha} - \alpha \rho S$$

Then, the radon content at the outlet can be obtained from the solution of this equation by taking  $x$  as  $vT'$  and satisfying the following boundary condition.

$$\begin{aligned} \rho &= \frac{\alpha CAh}{\alpha S\lambda + CA} \rho_\gamma \left\{ 1 - e^{-\left(\lambda + \frac{CA}{\alpha S}\right)(T-T')} \right\} \quad \text{at } x = 0. \\ \rho &= \frac{\alpha CAh}{\alpha S\lambda + Ca + Kb} \cdot \rho_\gamma \left\{ 1 - e^{-\left(\lambda + \frac{Ca}{\alpha S} + \frac{Kb}{\alpha S}\right)T'} \right\} \\ &+ \frac{\alpha CAh}{\alpha S\lambda + CA} \cdot \rho_\gamma \cdot e^{-\left(\lambda + \frac{Ca}{\alpha S} + \frac{Kb}{\alpha S}\right)T'} \left\{ 1 - e^{-\left(\lambda + \frac{CA}{\alpha S}\right)(T-T')} \right\} \end{aligned} \quad (4)$$

When the groundwater is perfectly surrounded by the rocks to its outlet, it may be expressed that  $b = 0$  and  $a = A$ . In order to find the highest value of  $\rho$  being possible for the constant value of  $\rho_\gamma$ , substituting into equ. (4) that  $b = 0$ ,  $a = A$  and  $T \rightarrow \infty$ ,

$$\rho = \frac{\alpha CAh}{\alpha S\lambda + CA} \rho_\gamma$$

In the extreme case of negligibly small  $CA$  compared with  $\alpha S\lambda$ , the upper limit of radon content of spring water discharging from such rocks is computed as follows.

$$\rho = \alpha h \rho_\gamma \quad (5)$$

$\alpha$  is the function of pressure, temperature and the kinds of solvent. Though it is expected to vary with the depth, we assume it for a simplification to be approximately constant.

Granite has about  $10^{-12}\text{g/g}$  of Ra concentration which is considered as the highest among those in commonly existing rocks. If we assume that the density of Granite is 2.5 and  $\alpha$  is 0.25 for  $20^\circ\text{C}$  of temperature and the standard atmospheric pressure, the highest Rn content possibly occurring in

the groundwater is found by equ. (5) as  $0.625 \times 10^{-12}$  curie/cc or  $6.25 \times 10^{-10}$  curie/l. Thus, this value is probably expected as the upper limit of radon content in the groundwater flowing through the region of no special radioactive mineral or spring sinter.

Most of the highest Ra concentrations of spring sinters ever discovered near the ground surface in the area of strongly radioactive springs are about  $10^{-9}$ g/g, and  $0.67 \times 10^{-9}$ g/g in Misasa is the highest.<sup>9)</sup> It is given by equ. (5) that the spring sinters of  $10^{-9}$ g/g Ra concentration, being uniformly distributed along the path of spring, cannot maintain the radon content of the spring as higher than  $6.25 \times 10^{-7}$ curie/l.

The highest Rn content ever found in Japan is  $4.36 \times 10^{-6}$ curie/l for No. A-9 Spring in Masutomi according to Kuroda<sup>1)</sup> and other few springs are known to contain higher than  $10^{-6}$ curie/l. Still higher contents would be possible if the loss of radon from the underground flow were perfectly eliminated in these springs.

Accordingly, it must be expected that there are more concentrated Ra-deposits under the ground than ever found on the ground surface, in order to maintain such high contents of radon. If the accumulation of Ra-deposit is limited in any finite region, still higher Ra concentration in them must be required. It is readily found from equ. (4) that the smaller the region of deposit, the higher is the concentration in it.

$$(b) \quad q \frac{\partial \rho}{\partial x} = \lambda h R - K b \frac{q}{\alpha} - \lambda \rho S$$

$$\text{Boundary condition,} \quad \rho = \frac{hR}{S} \left\{ 1 - e^{-\lambda(x-T)} \right\} \quad \text{at } x = 0.$$

Radon content at the outlet is obtained as follows,

$$\rho = \frac{\alpha \lambda h}{\alpha S \lambda + K b} R \left\{ 1 - e^{-\left(\lambda + \frac{K b}{\alpha S}\right) T'} \right\} + \frac{h}{S} R \cdot e^{-\left(\lambda + \frac{K b}{\alpha S}\right) T'} \left\{ 1 - e^{-\lambda(x-T)} \right\} \quad (6)$$

Substituting that  $b = 0$  and  $T \rightarrow \infty$  in order to gain the upper limit of  $\rho$ ,

$$\rho = R h / S$$

When the distribution of radium is limited in finite region where it is assumed that  $b = 0$ , the relation between the radon content of the discharging water and the total amount of radium is presented by equ. (3) in the last section.

When the Ra-deposit is accumulated only in such a shallow region that we can assume  $T = T'$ ,

$$\rho = \frac{hLR}{q} \frac{\alpha S \lambda}{\alpha S \lambda + kb} \frac{1 - e^{-\left(\lambda + \frac{Kb}{\alpha S}\right)T'}}{T'} \quad (7)$$

When the equ. (3) is applicable, the value of  $\frac{1 - e^{-\lambda T}}{T}$  varies so slightly with the change of  $T$  that it is practically proper to take it as constant  $1.2 \times 10^{-4}$  for the smaller  $T$  than one day as shown in the next Table.

$T(\text{min.})$	1	60	300	1440	2880	7200
$\frac{1 - e^{-\lambda T}}{T}$	1.2 $\times 10^{-4}$	1.2 $\times 10^{-4}$	1.2 $\times 10^{-4}$	1.15 $\times 10^{-4}$	1.05 $\times 10^{-4}$	0.83 $\times 10^{-4}$

Thus, in these cases, the total quantity of Ra which contributes to the amount of radon dissolving in the spring water, is found from the following simple formula.

$$LR = \frac{q\rho}{1.2 \times 10^{-4}h} \quad (\text{g}) \quad (8)$$

In the cases to which the equ. (7) can be applied, taking  $K$  approximately equal to the already mentioned value, some examples are calculated as follows.

$$\begin{aligned} \text{When } Kb/\alpha S = 10^{-2}, \quad T' = 60; \quad LR &= q\rho/0.75 \times 10^{-4}h \\ &T' = 300; \quad LR = q\rho/0.31 \times 10^{-4}h \\ \text{When } Kb/\alpha S = 5 \times 10^{-2}, \quad T' = 60; \quad LR &= q\rho/0.32 \times 10^{-4}h \\ &T' = 120; \quad LR = q\rho/0.17 \times 10^{-4}h \end{aligned}$$

Some discrepancies are shown from the results in equ. (8). It is found that this discrepancy becomes larger according to the increase of the value of  $Kb/\alpha S$  or  $T'$ .

As an example, if we assume that the total amount of radium supporting the largest Rn content of  $4.36 \times 10^{-6}$  curie/l in No. A-9 Spring in Masutomi is distributed under the state to which the equ. (8) is applicable, substituting  $q = 0.5$  1/min into (8),

$$LR = 0.0182 \quad (\text{g})$$

If it can be calculated from equ. (7) in which  $Kb/\alpha S = 10^{-2}$  and  $T' = 60$ ,

$$LR = 0.0291 \quad (\text{g})$$

In both cases, considerable amounts of radium are required.

#### §4. Total accumulated quantities of radium in sinters.

It is explained in the last section that higher radium concentration is



required in the underground spring sinters than ever discovered on the surface. Then, it is an essential problem to find whether such large amounts of radium to meet this requirement are accumulated or not, or how many parts of them contribute to the radon dissolving in radioactive springs if there are enough amounts of radium. Some hypothetical calculations are tried with a view to give a key for this problem using the data obtained by Kuroda and Yokoyama in Masutomi.<sup>10)</sup>

We assume the simple underground information in Masutomi Springs in which the difference between radium contents in each spring occurs only owing to that of deposition rate in it.

When it is assumed that radium content in spring water, being a g/l in the deep region, decreases to  $b$  g/l by only deposition until outlet, the accumulated amounts of radium along the course of spring are found to be increased with the rate of  $q(a-b)$  g/min. Let  $R$  be the accumulated amounts of radium per unit length. We can gain the next equation showing the variations in the total amounts of radium accumulated along the length of  $L$  cm.

$$\frac{dLR}{dt} = q(a-b) - \lambda' LR$$

Where  $\lambda'$  is the disintegration coefficient of radium which is equal to  $4.36 \times 10^{-4}$  year<sup>-1</sup>. Solving this equation under the condition of  $R=0$  at  $t=0$ ,

$$LR = \frac{q(a-b)}{\lambda'} (1 - e^{-\lambda' t})$$

For example, calculated values of  $LR$  for No. A-51 and A-49 Springs are shown as follows, assuming the highest radium content in Masutomi which is  $0.37 \times 10^{-13}$  g/cc as the value of  $a$ .

$T$ (year)		1	10	100	1000	10000
$LR$ (g)	A-49	$0.0356 \times 10^{-4}$	$0.0856 \times 10^{-3}$	$0.0747 \times 10^{-2}$	$0.0638 \times 10^{-1}$	0.0181
	A-51	$0.272 \times 10^{-4}$	$0.272 \times 10^{-3}$	$0.238 \times 10^{-2}$	$0.203 \times 10^{-1}$	0.0572

Radon contents in the spring waters, supposed to be maintained by such distributions of radium, are calculated by substituting these values of  $LR$  into equ. (3), and are shown in the next Table with the ratios,  $n$ , between the observed and above calculated radon contents.

$T$ (year)		1	10	100	1000	10000
A-49	$q\rho$ (curie/min)	$0.103 \times 10^{-8}$	$0.103 \times 10^{-7}$	$0.089 \times 10^{-6}$	$0.076 \times 10^{-5}$	$0.0217 \times 10^{-4}$
	$n$				1.76	0.58
A-51	$q\rho$ (curie/min)	$0.326 \times 10^{-8}$	$0.326 \times 10^{-7}$	$0.285 \times 10^{-6}$	$0.243 \times 10^{-5}$	$0.0685 \times 10^{-4}$
	$n$			1.16	0.146	0.048

If the present state of spring has been continued these  $10^4$  years or more so that the accumulated amounts of radium is now under nearly steady state, it is within the range of possibility to maintain the large content of radon in the spring by the decay of the accumulated radium in the deposits. But, though the highest value of  $\rho$  measured in each spring was used in these calculations, the value of  $n$  is probably expected to be much higher owing to the underground loss of radon as already mentioned.

Iwasaki<sup>2)</sup> showed in his experiments for the sample from Ikeda Springs that more than 60% of radon produced in the spring sinters could be dissolved into the water, but the correspondency between these experiments and the actual underground state is considered to be not clear.

Therefore, it must be expected for No. A-49 Spring that there is such a large radium concentration on the surface of the sinters that the radon produced in them is permitted to dissolve almost directly into the water or that the accumulated amounts of radium are still greater than those calculated.

## §5. Summary.

Assuming that the amounts of radon dissolved in the radioactive springs originate from radium involved in the mother rocks or spring sinters, analytical treatments were given to find the relation between radon content in the spring and radium concentration in the mother rocks and showed that the former cannot exceed the value of equilibrium with the latter even in the highest. In order to support the strong radioactivities of some springs, the larger radium concentration in the spring sinters than ever discovered must be expected under the ground.

The equations which permit the calculation of the total amounts of radium, contributing directly to radon in the spring water, were presented.

Outlines for the accumulated amounts of radium in the spring sinters were expressed concerning a few examples in Masutomi, and discussions were made with respect to the proportional amounts contributing to radon in the water to the total accumulated amounts of radium.

## §6, Acknowledgment.

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## 放射能泉に於けるラドンの源について (抄録)

吉 川 恭 三

温泉水中に溶解しているラドンがすべて、地下の岩石中のラヂウムから発生したものと考え、一様なラヂウム濃度をもつ岩石中を流動する水の含むべきラドン濃度に関する解析的研究が行われた。その結果、温泉水のラドン濃度は母岩のラヂウムと平衡にあるラドン濃度を超すことがないと結論される。実測せられている強放射能泉の幾つかのラドン濃度は之迄に知られている程度の沈澱物のラヂウム含有量からでは説明出来ず、地下に更に濃縮されたラヂウム沈澱物を予想しなければならない。地下に沈澱物として堆積されているラヂウムの量を計算する方法が呈出され、増富温泉での数例につき、その量と、其から発生したラドンの実際に温泉水に溶解している割合とが近似的に求められた。